IV.C.8 Enhanced Hydrogen Production Integration with Carbon Dioxide Separation in a Single-State Reactor

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Start Date: September 2003
Projected End Date: September 2005 (recently applied for one year no-cost extension)

Objectives

- Demonstrate the success of a technology to effectively and economically produce a pure hydrogen stream by coal gasification with integrated capture of CO_2 emissions, for subsequent sequestration.
- Demonstrate a high reactivity, mesoporous calcium sorbent for in-situ carbon dioxide separation.
- Test the regenerability of the sorbent over multiple calcination-carbonation cycles.

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Production section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

- L. Durability
- M. Impurities
- N. Defects
- O. Selectivity
- P. Operating Temperature
- Q. Flux
- S. Cost

Technical Targets

The primary target that the project will attempt to meet during its implementation is cost reduction of H_2 production from natural gas.

- \$3.00/gge H_2 (by 2005)
- \$1.50/gge H_2 (by 2010)

Approach

- Identify the temperature range selective towards carbonation by temperature-programmed reaction of the calcium oxide sorbent in a thermal gravimetric analyzer (TGA).
- Perform differential experiments for estimation of kinetic parameters of the carbonation reaction that will be followed by life cycle testing of the sorbent by in-situ carbonation and sub-atmospheric/steam calcination (regeneration).
- Carry out integral experiments to measure the effectiveness of commercial water-gas-shift (WGS) catalysts along with the calcium oxide sorbent for coupled WGS and carbonation reactions in the selected high temperature range.

Accomplishments

- The morphology of the calcium sorbent and the naturally occurring Linwood Hydrate sorbent were characterized.
- Completed thermodynamic analysis of the carbonation of CaO under simulated fuel gas conditions.
- Preliminary hydrogen generation experiments were conducted, the carbonation of CaO under multiple fuel gas concentrations was analyzed, and the initial catalyst screening was completed.

Future Directions

- Complete new reactor design and fabrication, including procurement and set-up of high temperature/high pressure magnetic suspension balance components. Complete new reactor shakedown/sorbent testing/ online gas analysis and evaluate the regeneration of CaS to provide a sulfur removal unit upstream of the carbonation/hydrogen generation reactor.
- Conduct multiple cycles of carbonation and calcination under simulated process conditions in the TGA. Complete hydrogen generation enhancements.

Introduction

The proposed research and development project examines an innovative, proprietary, CO_2 removal technique that can occur simultaneously with the production of hydrogen from fossil fuels.

Approach

The first step will involve identification of the temperature range that is selective towards carbonation by temperature-programmed reaction of the calcium oxide sorbent in a TGA. Differential experiments will be performed for estimation of kinetic parameters of the carbonation reaction. This will be followed by life cycle testing of the sorbent by in-situ carbonation and sub-atmospheric/steam calcination (regeneration). Subsequently, integral (breakthrough) experiments will be carried out to measure the effectiveness of commercial WGS catalysts along with the calcium oxide sorbent for coupled WGS and carbonation reactions in the selected high temperature range. Preliminary evaluation of the catalytic activity of calcium carbonate for WGS reaction alone by integral experiments will be followed by investigation of the catalytic activity of calcium for coupled WGS and carbonation reactions. Finally, the selectivity of CaO for H_2S/CO_2 by simultaneous heterogeneous gas-solid reactions will be explored.

Summary

The morphology of the calcium sorbent and the naturally occurring Linwood Hydrate sorbent were characterized. A thermodynamic analysis of the carbonation of CaO under simulated fuel gas conditions was also completed. Finally, preliminary hydrogen generation experiments were conducted; the carbonation of CaO under multiple fuel gas concentrations was analyzed; and the initial catalyst screening was concluded. An overall integration scheme of this process for hydrogen production from coal gasification is illustrated in Figure 1.

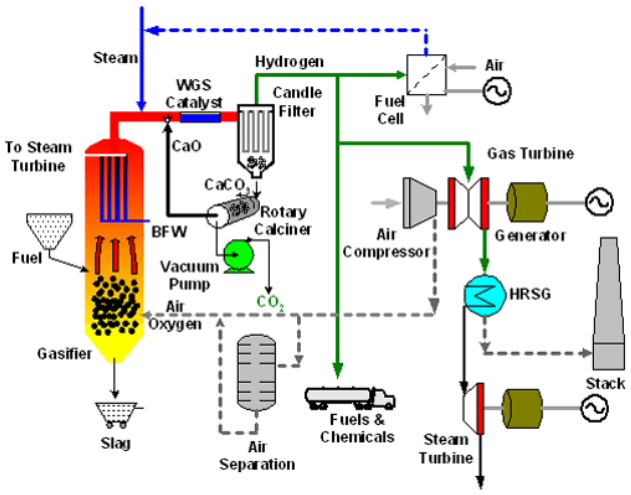

Figure 1. Hydrogen Production Process Schematic Using Carbonation of CaO to Remove CO_2

Special Recognitions & Awards/Patents Issued

1. "High Temperature CO_2 Capture with Enhanced Hydrogen Production using a Calcium Based Process." U.S. Provisional Patent Application (July 2005) (U.S. Patent Pending).

FY 2005 Presentations

1. "Enhancing Hydrogen Production With In-Situ CO_2 Separation Using CaO/Catalyst Systems" Iyer, M., Gupta, H., Sakadjian, B. and Fan, L-S. *AIChE Annual Tech. Meeting*, Austin, TX, 2004.
2. Gupta, H; Iyer, M.V.; Sakadjian, B.B.; and Fan, L.-S., "The Role of CaO in Maximizing Hydrogen Production from Fossil Fuels" Fuel Cell Seminar, San Antonio, TX, 2004

FY 2005 Publications

1. Gupta, H; Iyer, M.V.; Sakadjian, B.B.; and Fan, L.-S., "The Role of CaO in Maximizing Hydrogen Production from Fossil Fuels" Proceedings from Fuel Cell Seminar, San Antonio, TX, 2004